

## ACCUMULATION RATE AT MIZUHO STATION, EAST ANTARCTICA: AN APPLICATION OF Pb-210 METHOD

Noriyasu MASUDA and Koh HARADA

*Department of Chemistry, Faculty of Fisheries, Hokkaido University,  
1-1, Minatocho 3-chome, Hakodate 041*

**Abstract:** The lead 210 method for dating was applied to firn samples collected from a 5-m pit at Mizuho Station by the 15th Japanese Antarctic Research Expedition. Two series of measurements were undertaken (Po-210 and Pb-210) and the snow accumulation rates were estimated as  $19 \pm 2$  cm snow per year from the Po-210 profile and as  $20 \pm 2$  cm snow per year from the Pb-210 profile. The net accumulation rate was estimated as  $7.8 \pm 0.8$  g water per year assuming that the density was 0.4 g per cm<sup>3</sup>. These values well agreed with the results of previous studies. The lead 210 method could be applied to estimate snow accumulation rate within error of 10% as two sigma.

### 1. Introduction

Radon (Rn-222) is produced by the decay of Ra-226 in the crust and is released as gaseous phase from the earth's surface to the atmosphere. Rn-222 in the atmosphere is immediately decayed out (half life 2.8 days) to Pb-210 (half life 22 years) through some short-lived radionuclides. Pb-210 in the atmosphere is removed from the atmosphere with aerosol particles by dry and wet deposition. In the snow layer, Pb-210 is decayed constantly and if constant deposition of Pb-210 on the snow surface was assumed, accumulation rate of the snow can be estimated with the vertical profile of Pb-210 activities.

The lead 210 chronology was developed and applied to ice and firn samples from Antarctica and Greenland (CROZAZ *et al.*, 1964; CROZAZ and LANGWAY, 1966; PICCIOTTO *et al.*, 1967, 1971). Pb-210, a decay product of Rn-222, was used as a tracer radionuclide to estimate atmospheric particle flux to the earth's surface, and it also gives us time scale of natural phenomena.

Accumulation rate of Antarctic snow was usually estimated by the methods of gross beta-ray activity derived from nuclear bomb test (JOUZEL *et al.*, 1979; WATANABE, 1978),  $\delta^{18}\text{O}$  variation (DANSGAAD and JOHNSEN, 1969; WATANABE, 1978), snow stratigraphic interpretation (SHIMIZU, 1964; TAYLOR, 1971; WATANABE *et al.*, 1978), snow stakes (OKUHIRA and NARITA, 1978; YAMADA *et al.*, 1978), sodium profile (HERRON and LANGWAY, 1979; JHONSON, 1982) and solid microparticle profile (MARSHALL, 1962; THOMPSON *et al.*, 1981). However, the gross beta-ray method cannot trace the rate beyond a pre-nuclear test period. The  $\delta^{18}\text{O}$  profile may sometimes be affected and masked by the movement of water vapor in the snow on the formation of loose depth hoar. The

snow stakes method is the most accurate but it cannot be extended to the past. The methods of sodium and solid microparticle profiles could not identify the hiatus periods as well as the  $\delta^{18}\text{O}$  method.

The merits of the Pb-210 method are as follows: (1) Continuous tracing of the accumulation rate up to 200 years B.P. (2) Little is affected by the movement of water vapor in the snow layer because lead may not be present as gaseous phase. (3) Even if the surface snow layer was disturbed by wind, net accumulation rate can be estimated with a fixed layer below the disturbing layer. (4) Absolute age of the snow layer can be estimated. If the Pb-210 flux of the area was known, the age of snow layer can be traced back with Pb-210 decay. This method can be applied to snow samples including long-time hiatus or lack of snow surface due to wind. (5) Ice core including hiatus and accumulation rate change can be also applied. The hiatus will occur at various intervals. The accumulation rate is calculated from the gradient of isotopic ratio profiles in the Pb-210 method. If the ratio profile had discontinuity or change of its gradient, the hiatus period or accumulation rate change can be detected. PICCIOTTO *et al.* (1971) presented a typical hiatus-like profile of polonium isotopes (SPQMLT St. 3-5-248), but they did not mention hiatus episode. Furthermore, information about

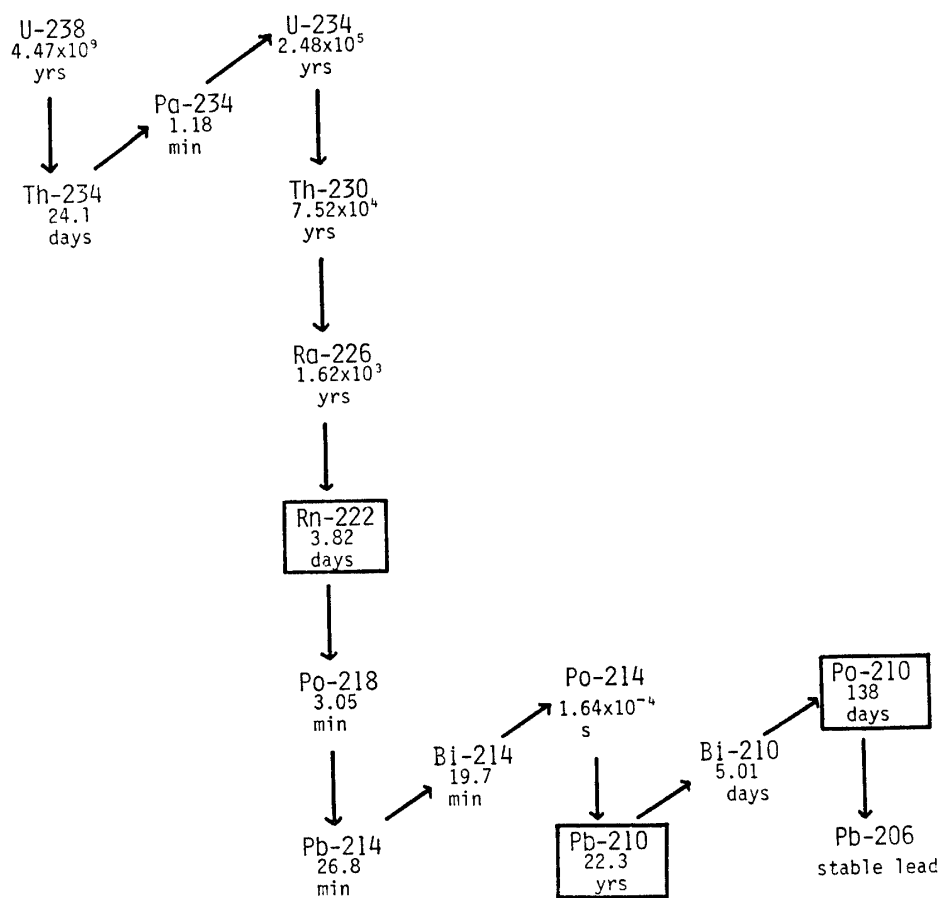


Fig. 1. The decay chain of the uranium 238 series isotopes and the half life of each isotope. Alpha decays are shown by the vertical arrows and beta decays by the diagonal arrows.

global aerosol transportation of crustal materials can be supplied. This information is not a purpose of this study, but it is helpful for geochemical understanding of elemental cycles on a global scale.

## 2. Method

Seven snow block samples (ca.  $45 \times 60 \times 30$  cm) were collected from the wall of a 5-m pit at Mizuho Station ( $70^\circ 42.1'S$ ,  $44^\circ 17.5'E$ , 2169 m a.s.l., WATANABE and YOSHIMURA, 1972) by the 15th Japanese Antarctic Research Expedition (JARE-15) and were stored frozen until analysis. The depths of samples are shown in Table 1. To determine the contents of Pb-210 and Po-210 in the snow, the modified method of FUKUDA and TSUNOGAI (1975) and HARADA and TSUNOGAI (1985) was used (Fig. 2). Radiochemical equilibrium was achieved between both radionuclides and Pb-210.

Outer 10 cm of each snow block was cut off to prevent contamination during storage, and the block was melted in the acid-washed polyethylene bag. The melt water

Table 1. Results of lead 210 measurements.

| Sample | Depth (cm) | Weight (kg) | Po-210 (counts) | Po-208 (counts) | $^{210}\text{Po}/^{208}\text{Po}$ (10 kg) | Pb-210 (cph/10 kg) |
|--------|------------|-------------|-----------------|-----------------|---|--------------------|
| M-1-2  | 0- 50      | 14.0        | 5004            | 1939            | $1.84 \pm 0.10$                           | nd                 |
| M-2-1  | 50-100     | 13.5        | 2209            | 1084            | $1.51 \pm 0.11$                           | nd                 |
| M-3-1  | 100-150    | 12.2        | 1818            | 813             | $1.98 \pm 0.16$                           | nd                 |
| M-4-2  | 150-200    | 14.0        | 2117            | 935             | $1.62 \pm 0.12$                           | nd                 |
| M-5-2  | 200-250    | 13.5        | 3699            | 1624            | $1.69 \pm 0.10$                           | $452 \pm 17$       |
| M-7-1  | 300-350    | 14.1        | 7429            | 3987            | $1.33 \pm 0.05$                           | $412 \pm 16$       |
| M-10-2 | 450-500    | 13.7        | 5106            | 3500            | $1.07 \pm 0.04$                           | $310 \pm 14$       |

The error denotes the counting statistics as  $2\sigma$ .

nd: not determined.

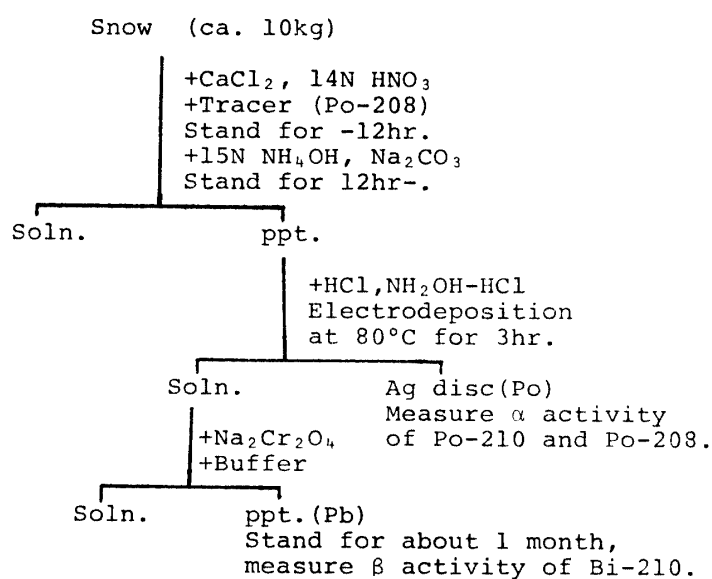


Fig. 2. The procedure for determination of Po-210 and Pb-210.

(12.2–14.1 kg) was added with 20 ml of 14N HNO<sub>3</sub>, a Po-208 tracer ( $8.00 \pm 0.57$  dpm), and 15 g of CaCl<sub>2</sub> as carrier, and then it was stirred and stood for 3 h. Lead and polonium were coprecipitated with calcium carbonate by adding 25 ml of 15N NH<sub>4</sub>OH and 45 g of Na<sub>2</sub>CO<sub>3</sub> and stood overnight. The precipitate was filtered and dissolved with HCl. A portion of NH<sub>2</sub>OH-HCl solution was added to the solution to mask ferric ion. Then, Po-210 and Po-208 were electrodeposited onto a silver disc at 80°C for 3–24 h. The solution remaining after the electrodeposition was added with Na<sub>2</sub>Cr<sub>2</sub>O<sub>4</sub> to cause lead chromate precipitation. The precipitation was filtered and stood for about one month to achieve the radioactive equilibrium between Bi-210 and Pb-210.

The activity of Po-210 and Po-208 on the silver disc was counted by using a surface barrier silicon detector coupled with a multichannel pulse-height analyzer for about 7000 min. The beta-ray activity on the chromate precipitate was measured by 2 $\pi$  gas-flow low background beta counter for 240 min.

The accumulation rate of snow was estimated with the vertical activity profiles of Pb-210 and Po-210. Po-208 is an artificial isotope and does not exist in natural environment.

### 3. Results and Discussion

Three snow samples were determined both of alpha activity (Po-210) and beta activity (Pb-210). The other four samples were determined only by Po-210 method. One of the polonium isotope results is shown in Fig. 3. The peak of Po-208 (5.113 Mev), a tracer, is presented in lower energy followed by the Po-210 peak (5.305 Mev). The data are presented in Table 1 and Fig. 4. The ratio of <sup>210</sup>Po/<sup>208</sup>Po was calculated with the sum of peak counts. The error was within 3.7–8% as two sigma.

The accumulation rate of snow at Mizuho Station was estimated from the gradients of isotope ratios (Fig. 4) as  $19 \pm 2$  cm snow per year with the Po-210 data and  $20 \pm 2$  cm snow per year with the Pb-210 data. If the density was assumed as 0.4 g per cm<sup>3</sup> at a layer shallower than 5 m, the accumulation rate was  $7.6 \pm 0.8$  g water per year from Po-210 and  $8.0 \pm 0.8$  g water per year from Pb-210, respectively.

WATANABE and YOSHIMURA (1972) preliminarily reported the annual snow accumulation rate at Mizuho Station as 390 cm in 10 years from the density profile and 390 cm in 12 years from the stratigraphic observation. And they suggested that the net accumulation rate was 14–15 cm per year and if a hiatus period was considered, the rate might be a half of the value (7.0–7.5 cm water per year). WATANABE *et al.* (1978) estimated the mean accumulation rate as 10.6 g/cm<sup>2</sup> per year by stratigraphic interpretation of the 150-m core from Mizuho Station. NARITA and MAENO (1979) also estimated the net accumulation rate at Mizuho Station as 7.0 g/cm<sup>2</sup> per year from the growth rate of crystal grains in snow above 35 m. The results of our study agreed well with their estimates. The accumulation rate of Antarctic snow could be estimated by the Pb-210 method within the error of 10%.

In the surface layer (<1.5 m deep), the polonium isotopic ratios were scattered (Table 1 and Fig. 4). One possible explanation may be snow drift formation caused by construction of new buildings at Mizuho Station. The other possibility is a constant perturbation of snow layer shallower than 1.5 m by wind in this region. The

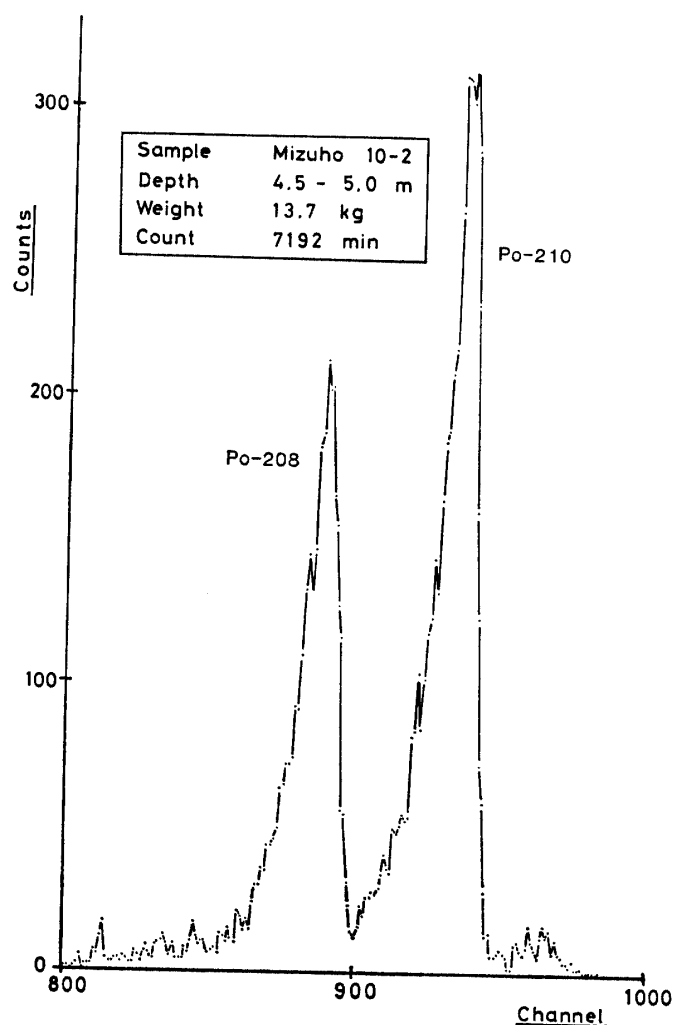


Fig. 3. Alpha-spectrum of Po-208 and Po-210.

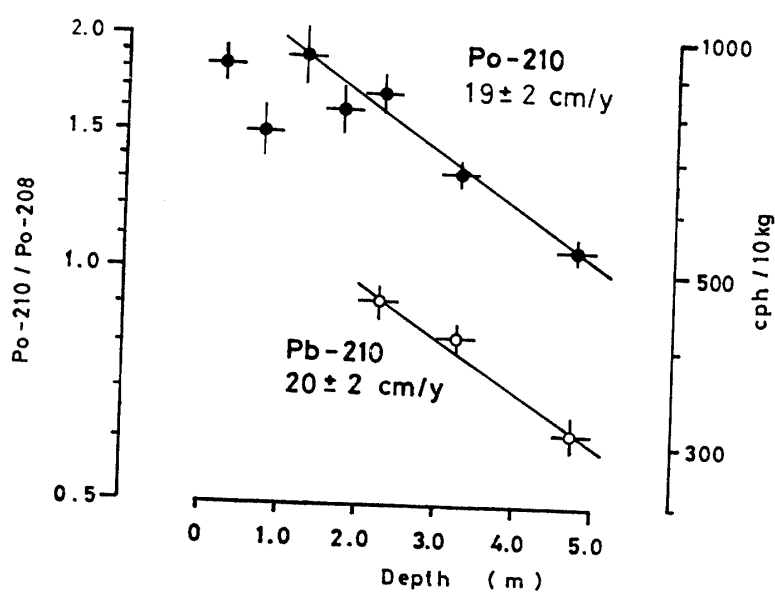


Fig. 4. The vertical profiles of Po-210/Po-208 activity ratio and Pb-210 (Bi-210) activity.

sampling conditions still remains as a possibility.

The following analysis should be undertaken for further understanding and application of the Pb-210 method to the Antarctic environment. (1) Detailed analysis with sampling at short intervals of depth to be applied to snow core including accumulation rate change and hiatus. Seasonal variation of crustal- and marine-originated material flux should be determined with the same core. (2) Large scale analysis covering a wide area and various elevations to estimate average accumulation rate. This may be helpful for water budget analysis and for clarifying the contribution of crustal material to the area. (3) Establishment of Pb-210 analysis with 0.5–1.0 kg snow samples. Usually the diameter of snow core samples available is 11 cm. The weight of snow core of one year accumulation is 760 g assuming that the accumulation rate was 20 cm per year and the density was 0.4. If the detailed dating for one year interval should be taken, analysis of small samples would be essential. To achieve the error within 10% as 2 sigma on 40 year old snow, two weeks counting should be taken. Furthermore, a clean room environment will be need for preparing samples.

### Acknowledgments

The authors wish to express their gratitude to Drs. M. SANO and T. IGARASHI for collecting samples, to Dr. K. WATANUKI for providing samples, and Drs. O. WATANABE and Y. FUJII for useful suggestions.

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*(Received March 24, 1986; Revised manuscript received June 10, 1986)*